

7 News from UK's CCP9 Programme

UK's Collaborative Computational Project 9 (CCP9) on "Computational Studies of the Electronic Structure of Solids"

7.1 Report on the CCP9 Conference

Daresbury Laboratory, UK

29-30 November 2004

<http://www.ccp9.ac.uk>

Organizers:

**James Annett (Bristol University) and Walter Temmerman (Daresbury
Laboratory)**

This conference, which was hosted at Daresbury Laboratory (UK), mostly brought together the different working groups of the British CCP9 Programme. Including external researchers from Japan, Germany and the USA, the conference was attended by 79 participants, many of them students or post-docs. The highlight of the conference was the Max Born Lecture and the Max Born Prize of the Institute of Physics (IoP) and the Deutsche Forschungs Gesellschaft (DFG), which was awarded to Matthias Scheffler (Fritz-Haber Institut, Berlin), for his outstanding achievements in theoretical surface physics, and in particular the combination of density functional theory and statistical mechanics to model adsorbates and catalytic processes.



Figure 1: Matthias Scheffler and John Beeby, the IoP representative

The conference started with a lecture by Richard Martin, in which he reviewed the developments of electronic structure theory over the last 40 years, leading to the current topics of the

field. This talk beautifully set the scene for the subsequent contributions, which reported on the current status of research in different branches of electronic structure theory, ranging from new theoretical developments to applications, allowing to make contact with experimentally accessible quantities. Amongst the theoretical developments concerning electronic correlations, there was the GW approximation. Rex Godby explained the method as such, including a detailed discussion on different levels of self-consistency, while Takao Kotani presented a novel "quasi-particle self-consistent" GW scheme, implemented in the FP-LMTO method, and first results on the band gaps for a number of semiconductors. Mike Towler reported on recent progress in the quantum Monte Carlo methods, highlighting some algorithmic developments, aimed at an all-electron treatment of heavy atoms and presented some applications using the 'CASINO' code. Dzigka Szotek discussed the electronic and magnetic properties of spintronics materials based on the self-interaction corrected local spin density approximation. A generalisation of density-functional theory for superconductors, allowing for first principles calculations of superconducting properties, including first applications to real systems, were presented by Martin Lüders. Another methodological development was the non-local extension of the coherent potential approximation and its implementation within the KKR method by Derwyn Rowlands. Mikhail Katsnelson discussed the effect of incoherent non-quasiparticle states in half-metallic ferromagnets leading to their anomalous magnetic and electronic properties. A central theme of the conference was concerned with the realistic description of large systems. This field was opened by the Max Born lecture, given by Matthias Scheffler, in which he gave an overview of the applied methods and discussed the example of O₂ dissociation on Al(111). Other contributions in this area were the plenary talk of Mike Payne, who introduced two new methods, namely a linear scaling code and a hybrid modelling scheme, which will allow for the calculation of tens of thousands atoms over long time scales. The linear scaling code was described in more detail by Chris-Kriton Skylaris. A different approach to large systems was outlined by David Pettifor, who described the method of constructing analytical bond-order potentials from systematically coarse-graining DFT results via an intermediate tight-binding model. Bob Jones reported on DFT calculations of extended interstitial defects in Si and demonstrated that these complex arrangements of interstitials can be analysed theoretically. A method for predicting magnetic resonance experiments, based on DFT calculations was put forward by Chris Pickard. He also described how the method is used by the experimental community to assist in extracting information from the experimental data.

Apart from the talks there were poster-sessions during the buffet lunches, and a bottle of champagne was awarded to Jan Minar (Munich) for the best poster presentation.

The conference contained a good mixture of overview talks and more specialised contributions. Throughout the whole conference the atmosphere was very lively, and many stimulating discussions took place. To this good atmosphere also contributed the banquet dinner at the Yang Sing restaurant in Manchester, which was held in honour of Matthias Scheffler on the occasion of his Max Born Prize.

The photographs of the conference can be found on the web under the URL:

<http://www.ccp9.ac.uk/data/CCP9WorkshopPhotos/index.htm>

Martin Lüders, *Daresbury Laboratory*

Programme of the CCP9 Conference 2004

Monday, 29th November

09.30 - 10.30		Registration and coffee
10.30 - 11.30	Richard Martin	Building on the successes of Electronic Structure Theory
11.30 - 11.55	Mike Towler	Recent progress in quantum Monte Carlo
12.00 - 12.25	Dzidka Szotek	Self-interaction corrected Local Spin Density calculations for spintronics materials
12.30 - 14.30		Buffet Lunch and Posters
14.30 - 15.30	Rex Godby	The GW approach to spectral, ground-state and transport properties
15.30 - 15.55	Bob Jones	Density functional treatment of extended interstitial defects in Si
16.00 - 16.25	Martin Lüders	Ab-initio theory of superconductivity: Density functional formalism, approximate functionals and first applications
16.30 - 17.00		Coffee
17.00 - 18.00	Born Prize Presented to Matthias Scheffler	Statistical Mechanics from First Principles
20.00		Conference Dinner

Tuesday, 30th November

08.45		Coffee and Pastries
09.00 - 10.00	Mike Payne	Next Generation Simulation Techniques
10.00 - 10.25	Chris-Kriton Skylaris	Introducing ONETEP: Linear-Scaling Density Functional Theory with Plane Waves
10.30 - 10.55	David Pettifor	Recent Developments in analytic bond-order potentials
11.00 - 11.30		Coffee
11.30 - 11.55	Derwyn Rowlands	First-principles description of disordered metallic systems with short-range order
12.00 - 12.25	Chris Pickard	Interpreting magnetic resonance experiments from first principles
12.30 - 14.55		Lunch and posters
15.00 - 16.00	Mikhail Katsnelson	Non-quasiparticle states in half-metallic ferromagnets
16.00 - 17.00	Takao Kotani	Quasiparticle self-consistent GW method

List of Participants

Mahrous Ahmed, University of Sheffield, php02mra@shef.ac.uk
James Annett, University of Bristol, james.annett@bristol.ac.uk
Gary Batt, University of Bristol, GB0657@bris.ac.uk
John Beeby, University of Leicester, zjb@le.ac.uk
Richard Blake, CCLRC Daresbury Laboratory, r.j.blake@dl.ac.uk
Nuno Carneiro, University of York, nfc103@york.ac.uk
Roger Cavallaro, University of Salford, r.cavallaro@pgr.salford.ac.uk
Rathin Choudhury, University College London, r.choudhury@ucl.ac.uk
Stewart Clark, University of Durham, s.j.clark@durham.ac.uk
Simon Crook, EPSRC, Simon.Crook@epsrc.ac.uk
Gabor Csanyi, University of Cambridge, gc121@cam.ac.uk
Markus Daene, Martin-Luther-University Halle-Wittenberg, daene@physik.uni-halle.de
Peter H. Dederichs, Research Center Juelich, p.h.dederichs@fz-juelich.de
Andy Duff, University of Bristol, A.I.Duff@bristol.ac.uk
Stephen Dugdale, University of Bristol, s.b.dugdale@bristol.ac.uk
Paul Durham, CCLRC Daresbury Laboratory, p.j.durham@dl.ac.uk
Hubert Ebert, University of Munich, Hubert.Ebert@cup.uni-muenchen.de
Arthur Ernst, MPI Halle, aernst@mpi-halle.de
Barbara Falabretti, University of Cambridge, bf222@cam.ac.uk
Jacob Gavartin, University College London, j.gavartin@ucl.ac.uk
Gillian Gehring, University of Sheffield, g.gehring@shef.ac.uk
Michael Gibson, University of Durham, m.c.gibson@durham.ac.uk
Nikitas Gidopoulos, CCLRC Rutherford Appleton Lab, N.Gidopoulos@rl.ac.uk
Mike Gillan, University College London, m.gillan@ucl.ac.uk
Rex Godby, University of York, rwg3@york.ac.uk
Balazs Gyorffy, University of Bristol, b.gyorffy@bristol.ac.uk
Volker Heine, University of Cambridge, vh200@cam.ac.uk
Ian Hughes, University of Warwick, I.D.Hughes@warwick.ac.uk
Dominik Jochym, University of Durham, dominik.jochym@durham.ac.uk
Robert Jones, University of Exeter, r.jones@exeter.ac.uk
Mikhail Katsnelson, Radboud Universiteit Nijmegen, M.Katsnelson@science.ru.nl
Samantha Keil, , mdt26@cam.ac.uk
Diemo Koedderitzsch, Martin-Luther-Universitt Halle-Wittenberg, d.koedderitzsch@physik.uni-halle.de
Takao Kotani, Osaka University, kotani@phys.sci.osaka-u.ac.jp
Seung Cheol Lee, CCLRC Daresbury Laboratory, scle@dl.ac.uk
Seung Mi Lee, University of Oxford, seung.lee@materials.ox.ac.uk
Andrew Leung, City University of Hong Kong, andrew.leung@cityu.edu.hk

Weixing Li, University of Salford, W.Li@pgr.salford.ac.uk
Martin Lueders, CCLRC Daresbury Laboratory, m.lueders@dl.ac.uk
Andrea Ma, University of Cambridge, am570@cam.ac.uk
Richard Martin, University of Illinois, RMartin@uiuc.edu
Jan Minar, University of Munich, jan.minar@cup.uni-muenchen.de
Carla Molteni, King's College London, carla.molteni@kcl.ac.uk
Ian Morrison, University of Salford, i.morrison@salford.ac.uk
Ashkan Naeini, Siemens, ashkan.naeini@siemens.com
Tsogbadrakh Namsrai, University of Salford, T.Namsrai@pgr.salford.ac.uk
Duc Nguyen-Manh, UKAEA Culham Division, duc.nguyen@ukaea.org.uk
Kazuyoshi Ogasawara, Kwansai Gakuin University, ogasawara@ksc.kwansei.ac.jp
Sergey Ostanin, University College London, S.Ostanin@warwick.ac.uk
Sergiusz Osuchowski, University of Salford, serrz@agh.edu.pl
Mike Payne, University of Cambridge, mcp1@cam.ac.uk
David Pettifor, University of Oxford, david.pettifor@materials.ox.ac.uk
Chris Pickard, University of Cambridge, cjp20@cam.ac.uk
Emma Podnieks, University of Bristol, emma.podnieks@bristol.ac.uk
Keith Refson, CCLRC Daresbury Laboratory, K.Refson@rl.ac.uk
Duncan Riley, University of Salford, d.j.riley@pgr.salford.ac.uk
Ben Ringham, Loughborough University, B.D.Ringham@lboro.ac.uk
Derwyn Rowlands, University of Bristol, d.a.rowlands@bristol.ac.uk
Benedict Russell, University of Cambridge, bjr27@cam.ac.uk
Vicente Salinero, University of Durham, v.t.salinero@durham.ac.uk
Matthias Scheffler, Fritz-Haber-Institut der Max-Planck-Gesellschaft, scheffler@FHI-Berlin.mpg.de
Uwe Schonberger, University of Oxford, uwe.schonberger@materials.ox.ac.uk
Chris-Kriton Skylaris, University of Oxford, cks22@cam.ac.uk
Martin Stankovski, University of York, ms519@york.ac.uk
Julie Staunton, University of Warwick, j.b.staunton@warwick.ac.uk
Malcolm Stocks, Oak Ridge National Laboratory, gms@ornl.gov
Paul Strange, Keele University, P.Strange@phys.keele.ac.uk
Zdzislawa Szotek, CCLRC Daresbury Laboratory, z.szotek@dl.ac.uk
Ferenc Tasnadi, IFW Dresden, f.tasnadi@ifw-dresden.de
Walter Temmerman, CCLRC Daresbury Laboratory, w.m.temmerman@dl.ac.uk
Milica Todorovic, University of Oxford, milica.todorovic@materials.ox.ac.uk
Mike Towler, University of Cambridge, mdt26@cam.ac.uk
Paul Tulip, University of Warwick, P.R.Tulip@warwick.ac.uk
Balazs Ujfalussy, Research Inst. Solid State Physics, Budapest, bu@szfki.hu
Younsuk Yun, Kyung Hee University, ysyun@kriss.re.kr
Bernard Zarychta, Keele University, B.Zarychta@phys.keele.ac.uk
Hand Zenia, University of Sheffield, H.Zenia@sheffield.ac.uk
Guang Zheng, University of Durham, guang.zheng@durham.ac.uk

ABSTRACTS:

Talks

Building on the Successes of Electronic Structure Theory

Richard M. Martin

Department of Physics University of Illinois 1110 W. Green St. Urbana, IL 61801 USA

Understanding of electronic structure of materials has come a long way in the 80 years since 1924 when Prince Louis de Broglie deposited his thesis. From the start electrons have played a key role in the development of quantum theory, and within a few years quantum mechanics provided the underpinnings of present understanding of metals, insulators and semiconductors. Today the field is at a momentous stage, with new algorithms and computational methods, and rapid advances in basic theory. In the 40 years since the advent of density functional theory and the 20 years since the landmark Car-Parrinello paper, the work of many people have brought the field to the point where properties of materials can now be determined directly from the fundamental equations for the electrons [1]. The methods have become standard tools - an essential part of modern materials research. At the same time there have been important developments in many-body theory and methods to treat excitations of correlated, interacting electrons.

This talk is oriented toward the future and the greatest challenges, which are to understand the vast array of phenomena exhibited by the many-body system of interacting electrons in matter. At this point in time there is the opportunity to build upon recent successes to create new approaches that will make possible robust predictions for new phenomena and materials, complex systems, nanostructures, activity of large molecules in solution, magnetism, metal-insulator transitions, transport, and many other areas. Specific examples of recent work show the power of combined independent-particle and many-body methods to provide new predictive capabilities and new insights into important problems in physics, chemistry, and materials science.

[1] An exposition of the basic theory and methods along with extensive references and examples of recent work can be found in "Electronic Structure: Basic theory and practical methods," R. M. Martin, Cambridge University Press (2004), and on-line at <http://ElectronicStructure.org>.

Recent progress in quantum Monte Carlo

Mike Towler

Theory of Condensed Matter, University of Cambridge, UK

Abstract

In this talk I shall describe certain algorithmic developments in diffusion Monte Carlo simulations which lead to improved scaling with system size and an increased ability to treat heavy atoms without using pseudopotentials. Some applications of this highly-accurate technique using the code 'CASINO' will be outlined.

Self-Interaction Corrected Local Spin Density Calculations for Spintronics Materials

Z. Szotek

Computational Science and Engineering Department, Daresbury Laboratory, UK

Abstract

Self-interaction corrected (SIC) local spin density (LSD) method is applied to study electronic structure and magnetic properties of III-V and II-VI diluted magnetic semiconductors, magnetite, and such spinel ferromagnetic insulators as NiFe₂O₄ and CoFe₂O₄. In the diluted magnetic semiconductors we concentrate on the understanding and realization of carrier mediated ferromagnetism. The insulating charge ordered Verwey phase of magnetite is discussed in relation to the electronic structure of NiFe₂O₄ and CoFe₂O₄, and we speculate on the implications for possible technological applications.

The GW approach to spectral, ground-state and transport properties

Rex Godby

*Department of Physics, University of York,
Heslington, York YO10 5DD, United Kingdom*

Abstract

The GW approximation is the first-order term in an expansion of the self-energy operator Σ in powers of the dynamically screened electron-electron interaction, W . It may be thought of as exact exchange (the Hartree-Fock exchange operator), together with an partial inclusion of correlation diagrams through dynamical screening of the Coulomb interaction. From Σ , the one-electron Green's function G may be calculated, from which various spectral and ground-state properties are available. For transport properties (as well as certain other excited-state properties), the two-electron Green's function is required, which may also be formulated at the same level of approximation as GW .

I shall start with a brief review of key applications of GW to spectral properties such as quasiparticle energies and lifetimes for added electrons or holes, including some recent work on metallic clusters [1].

In applying GW to the ground-state total energy, the choice of whether G and/or W are made to be consistent with the Green's function that arises *from* Σ is particularly important: G_0W_0 , GW_0 and fully self-consistent GW , where G_0 generally indicates the LDA Green's function. I shall present results for finite and infinite systems at all three levels of self-consistency. These include converged results resulting from the incorporation of self-consistency and GW total-energy techniques into our general-purpose GWST "space-time" supercell code suite [2], which interfaces with pseudopotential plane-wave DFT calculations as its input. This allows the extension of the GW total-energy approach from high-symmetry test systems [3] to general systems, and opens the possibility of applications to systems whose energetics is not described sufficiently reliably within the usual DFT-based approaches.

I shall also describe our recent formulation [4] of the conductance of a junction between two nanowires in a form which permits the inclusion of electronic correlation effects in a GW -like framework.

- [1] "Image States in Metal Clusters", Patrick Rinke, Kris Delaney, P. García-González and R.W. Godby, submitted
- [2] "Space-time method for *ab initio* calculations of self-energies and dielectric response functions of solids", H. N. Rojas, R. W. Godby and R. J. Needs, Phys. Rev. Lett. **74** 1827 (1995)
- [3] "Many-body GW calculations of ground-state properties: quasi-2D electron systems and van der Waals forces", P. García-González and R. W. Godby, Phys. Rev. Lett. **88** 056406 (2002)
- [4] "Conductance and polarization in quantum junctions", P. Bokes and R.W. Godby, Phys. Rev. B **69** 245420 (2004)

Further details at <http://www-users.york.ac.uk/~rwg3/>.

Density functional treatment of extended interstitial defects in Si

R. Jones¹, T. A. G. Eberlein¹, J. P. Goss², P. R. Briddon²,
A. T. Blumenau³, S. Öberg⁴

¹*School of Physics, University of Exeter, Exeter EX4 4QL, U.K*

²*School of Natural Science, Newcastle upon Tyne NE1 7RU, U.K.*

³*Theoretische Physik, Universität Paderborn, 33098 Paderborn, Germany*

⁴*Department of Mathematics, Luleå University of Technology,
SE-97187 Luleå, Sweden*

Abstract

Self-interstitials in silicon, produced by ion-implantation or oxygen precipitation, can aggregate to form extended rod-like-defects (RLDs) with unusual $\{113\}$ habit planes and having both electrical and optical activity. Although known for over 30 years they remain an enigma. Here, we describe the results of local density functional calculations on the RLDs which reveal their structure and electrical activity. We find that small $\{113\}$ RLDs are more stable than RLDs condensing onto $\{111\}$ planes but this reverses for larger defects. We

attribute the electrical activity of $\{113\}$ RLDs to the bounding dislocations and a pronounced and sharp photoluminescence band to vacant interstitial sites on the RLD. The results of the modelling demonstrate that complex arrangements of interstitials are amenable to theoretical analysis and give hope that the pathways to other extended defects can also be found.

Ab-initio theory of superconductivity: Density functional formalism, approximate functionals and first applications

M. Lüders^{1,2}, M. A. L. Marques^{3,2}, N. N. Lathiotakis^{3,2}, A. Floris^{3,4},
G. Profeta⁵, L. Fast^{6,2}, A. Continenza⁵, S. Massidda⁴, E. K. U. Gross^{3,2}

¹*Daresbury Laboratory, Warrington WA4 4AD, United Kingdom*

²*Universität Würzburg, Am Hubland, D-97074 Würzburg, Germany*

³*Freie Universität Berlin, Arnimallee 14, D-14195 Berlin, Germany*

⁴*INFM SLACS, and Università degli Studi di Cagliari,
I-09124 Monserrato (Cagliari), Italy*

⁵*C.A.S.T.I. - Istituto Nazionale Fisica della Materia (INFM) and
Università degli studi dell'Aquila, I-67010 Coppito (L'Aquila) Italy*

⁶*SP Swedish National Testing and Research Institute, S-501 15 Borås, Sweden*

Abstract

A novel approach to the description of superconductors in thermal equilibrium is developed within a formally exact density-functional framework. The theory is formulated in terms of three “densities”: the ordinary electron density, the superconducting order parameter, and the diagonal of the nuclear N -body density matrix. The electron density and the order parameter are determined by Kohn-Sham equations that resemble the Bogoliubov-de Gennes equations. The nuclear density matrix follows from a Schrödinger equation with an effective N -body interaction. These equations are coupled to each other via exchange-correlation potentials which are universal functionals of the three densities. Approximations of these exchange-correlation functionals are derived using the diagrammatic techniques of many-body perturbation theory. The bare Coulomb repulsion between the electrons and the electron-phonon interaction enter this perturbative treatment on the same footing. In this way, a truly ab-initio description is achieved which does not contain any empirical parameters.

This new formalism is first applied to simple metals, to show the applicability to both weak and strong electron-phonon coupling. Further results for MgB_2 , and Al and Li under pressure will be presented.

Statistical Mechanics from First Principles

Matthias Scheffler

Fritz-Haber-Institut der Max-Planck-Gesellschaft

Faradayweg 4-6, D-14195 Berlin, Germany

Scheffler@FHI-Berlin.mpg.de

<http://www.fhi-berlin.mpg.de/th/th.html>

Abstract

This talk sketches the methods and describes recent theoretical work performed with *ab initio* atomistic thermodynamics and *ab initio* statistical mechanics (molecular dynamics as well as kinetic Monte Carlo).

Key examples in this talk concern the puzzle of O₂ dissociation at Al(111), in particular emphasizing the role of the O₂ spin state, and the properties of transition metals in oxygen atmospheres with (T, p) settings ranging from UHV to those encountered at “human” (or higher T, p) conditions.

Analyzing oxidation catalysis at the late transition metals it is found that the surface composition, that is attained under reactive conditions, and the consequential functionality of the surface can be very different from what is found at (or close to) thermodynamic equilibrium.

Next Generation Simulation Techniques

Mike Payne

TCM Group, Cavendish Laboratory, Madingley Road, Cambridge, CB3 0HE, UK

Abstract

In less than twenty years, the plane wave pseudopotential technique has progressed from modelling two atoms of silicon to modelling many hundreds of atoms of any species. However, this technique is still far away, both in terms of accessible lengthscale and more importantly accessible timescale, from being able to address many scientific challenges. In this talk I shall discuss two new techniques which confront these lengthscale and timescale challenges. They are a linear scaling first principles total energy technique, which will be described in greater detail in the talk by Dr. Skylaris, and a hybrid modelling scheme, developed in collaboration with Dr. Sandro De Vita. The linear scaling code provides the same accuracy as the plane wave pseudopotential technique but can be applied to systems containing many thousands of atoms, with the prospect of tens of thousands of atoms in the near future. The hybrid modelling scheme provides the same accuracy as a full quantum mechanical calculation but

allows simulations to be performed on systems containing hundreds of thousands of atoms or more for very long simulation times.

Introducing ONETEP: Linear-Scaling Density Functional Theory with Plane Waves

Chris-Kriton Skylaris

Department of Physical & Theoretical Chemistry,
South Parks Road, Oxford OX1 3QZ

Peter D. Haynes, Arash A. Mostofi and Mike C. Payne
Theory of Condensed Matter group, Cavendish Laboratory,
Madingley Road, Cambridge CB3 0HE

In order to apply first principles quantum mechanical calculations to systems containing thousands of atoms we need linear-scaling techniques. In achieving this goal we must not lose the accuracy, efficiency and flexibility provided by conventional total energy schemes. Plane waves are a popular choice of basis set for many such schemes because their implementation is straightforward and the completeness of the basis set can be controlled systematically with a single parameter. However, the resulting calculations require a computational effort which scales as the cube of the number of atoms in the system, and as a result current simulations are limited to hundreds of atoms.

In this talk I will present ONETEP (Order-N Electronic Total Energy Package), a parallel implementation of our linear-scaling reformulation [1] of the plane wave pseudopotential approach. The linear-scaling is achieved by exploiting the real space localisation of the electronic system that is inherent in non-metallic materials. I will describe the theoretical developments that allow us to optimise with linear cost the strictly localised quantities expressed in terms of a delocalised plane-wave basis. These same localised quantities also lead us to a physical way of dividing the computational effort among many processors to allow calculations to be performed efficiently on parallel machines. It will be shown that the localisation is achieved in a controlled and mathematically consistent manner so that ONETEP provides the same accuracy as conventional cubic-scaling plane-wave approaches, and offers fast and stable convergence. The particular features of our method which result in its success will be highlighted and preliminary results for realistic biological and condensed matter applications will be presented.

[1] C.-K. Skylaris, A. A. Mostofi, P. D. Haynes, O. Diéguez and M. C. Payne, *Phys. Rev. B*, **66**, 035119 (2002).

Analytic bond-order potentials for modeling the growth of semiconductor films

D.G. Pettifor¹, R. Drautz¹, D. Nguyen-Manh², D.A. Murdick³,
X.W. Zhou³, and H.N.G. Wadley³

¹*Department of Materials, University of Oxford*

²*UKAEA Fusion, Culham Science Centre*

³*Department of Materials Science and Engineering, University of Virginia*

Abstract

Robust interatomic potentials for simulating the growth of semiconductor films should be able to describe bond breaking and remaking naturally within their remit. In this paper we outline the derivation of such potentials, the so-called bond-order potentials (BOPs), by performing a systematic coarse graining from the electronic to atomistic modeling hierarchies. First the density functional theory (DFT) electronic structure and binding energy is simplified by introducing the tight-binding (TB) bond model whose parameters are determined directly from the DFT results. Second, this TB electronic structure is coarse grained through atom-centered moments and bondcentered interference paths, thereby predicting the analytic form of the interatomic BOP. We show that these interatomic potentials predict structural energy differences in quantitative agreement with k-space TB calculations and reproduce the experimental structural trends across the sp-valent elements.

First-principles description of disordered metallic systems with short-range order

Derwyn Rowlands¹, Julie Staunton², Balazs Györffy¹,
Ezio Bruno³, Beniamino Ginatempo³

¹*H.H. Wills Physics Laboratory, University of Bristol,
Bristol BS8 1TL, U.K.*

²*Dept. of Physics, University of Warwick, Coventry CV4 7AL, U.K.*

³*Dipartimento di Fisica, Università di Messina Salita Sperone 31,
98166 Messina, Italy*

Abstract

For many years the coherent potential approximation (CPA) has been widely used to describe the electronic structure of disordered systems. However, as a local mean-field theory of disorder, the CPA is unable to describe important environmental effects such as short-range order in alloys and spin fluctuations in magnets, amongst others. Only recently,

the first satisfactory means of going beyond the CPA has been developed, the non-local coherent potential approximation (NLCPA),[1,2] which has also been derived within the first-principles Korringa-Kohn-Rostoker (KKR) multiple-scattering framework.[3] In this talk I describe the method and illustrate with KKR-NLCPA calculations for the effects of short-range order upon the electronic structure of alloys such as *CuZn* and *CuNi*. [4] I also discuss future applications such as transport properties, and developments such as combination with density functional theory for ab-initio calculations, which will enable effects such as charge correlations and lattice displacements to be taken into account self-consistently for the first time.

- [1] M. Jarrell and H.R. Krishnamurthy, PRB 63, 125109 (2001)
- [2] R. Moradian, B.L. Gyorffy, and J.F. Annett, PRL 89, 287002 (2002)
- [3] D.A. Rowlands, J.B. Staunton, and B.L. Gyorffy, PRB 67, 115109 (2003)
- [4] D.A. Rowlands, J.B. Staunton, B.L. Gyorffy, E. Bruno, B. Ginatempo (to be published)

Interpreting magnetic resonance experiments from first principles

Chris J. Pickard, Jonathan Yates, and Francesco Mauri

Abstract

A method for predicting the results of magnetic resonance (NMR/EPR) experiments will be presented. It is based on the plane wave pseudopotential method, within density functional theory. Using a modified (gauge including) version of the projector augmented wave method, all-electron accuracy can be achieved. I will describe how the method is being adopted by the experimental community to assist in the extraction of the maximum amount of information from their magnetic resonance experiments. The range of applicability of the method will be emphasized, by reference to successful collaborative applications: porphyrin molecules, boron carbides, complex zeolite silicates, zircon, sodium and calcium silicate glasses and hydrogen bonded molecular crystals.

Non-quasiparticle states in half-metallic ferromagnets

Mikhail Katsnelson

University of Nijmegen, The Netherlands

Abstract

Anomalous magnetic and electronic properties of the half-metallic ferromagnets (HMF) are discussed. A general concept of the HMF electronic structure which takes into account

the most important correlation effects from electron-magnon interactions, that is, the spin-polaron effects, is presented. Special attention is paid to the so called non-quasiparticle (NQP), or incoherent, states which are present in the gap near the Fermi level. Manifestations of NQP in electronic density of states, tunneling transport, nuclear magnetic relaxation, core-level spectra and other properties of HMF are considered. First-principle calculations of the NQP- states for various half-metallic ferromagnets within the local-density approximation plus dynamical mean field theory (LDA+DMFT) are reviewed.

Quasiparticle self-consistent GW

Takao Kotani,²Mark van Schilfgaarde³Sergey V. Faleev, and⁴Takashi Miyake

Osaka University, Toyonaka 560, Japan

²*Arizona State University, Tempe, AZ, 85284*

³*Sandia National Laboratories, Livermore, CA 94551*

⁴*Tokyo Institute of Technology, Tokyo 152-8551, Japan*

We present some progress in the development of a semi self-consistent GW method based on the all-electron, full-potential LMTO method. Starting from a trial one-body Hamiltonian H_0 (or bare Green function G_0), we can get the self-energy $\Sigma(\omega)$ and also the Green function G in the usual GW calculation. Then we can define new one-body Hamiltonian H'_0 by fixing the energy-dependence of $\Sigma(\omega)$ (we tested some possibilities), so that H'_0 can well reproduce the quasiparticle (QP) part contained in G . Here we require $H_0 = H'_0$, so as to determine H_0 . This is a self-consistency requirement taken as “**non-interacting Hamiltonian \approx QP part of G** ”. Thus we call our method as QPsc GW . This is a kind of self-consistent perturbation theory. This H_0 gives “the *best* bare QP picture”, which get to be the independent-particle picture for the weakly-correlated systems. Then QPsc GW gives directly the QP density of states (QP-DOS) in the sense of independent-particle picture, which can be justified by the Landau’s Fermi liquid theory. The QP bands given by H_0 can be used directly within the independent-particle picture to evaluate quantities, e.g. response functions, transport, and so on.

The “bare QP picture” should be also a rigid starting point even when we try to go beyond the independent-particle picture. The method may be viewed as a way construct the most suitable quadratic part of the full Hamiltonian for many-body perturbation theory. As for the spectrum DOS (SP-DOS), the imaginary part of the full Green function G , it can be calculated by a one-shot GW calculation (or GW +extensions) from the QPsc GW result.

This situation is in contrast with the full self-consistent GW method (full sc GW), which makes SP-DOS self-consistent. Compared the full sc GW , QPsc GW is advantageous in two ways:

- (1) Numerically easier and more stable. We successfully applied it to wide range of materials.
- (2) There is self-consistency in both the QP bands and the dynamically screened Coulomb interaction W (they are obtained at the same time). By contrast, the full sc GW give problematic W , which results in poor G .

We applied our QPsc GW to metals, semiconductors, oxides, magnetic materials, f -electron materials (in progress) and so on. Our results show very systematic improvement over LDA

compared with experiments. We present two key findings:

- (1) large improvements are found for materials where one-shot GW fails.
- (2) The discrepancies with experiment are very systematic, and can be explained in terms of what GW theory omits from the exact hamiltonian.

As for semiconductors, not only band gaps but many other properties are improved, such as effective mass. For NiO and MnO, $d-d$ splitting and relative positions of oxygen $2p$ bands move closer to experimental results. Transition metals show a small, systematic d -band width narrowing relative to LDA. We also present recent results for MnGaAs and half-metallic compounds.

Finally, we report some progress on attempts to compute the total energy in RPA without the LDA kinds of approximations.

[1] Sergey V. Faleev, Mark van Schilfgaarde, and Takao Kotani, PRL in press. cond-mat/0310677

[2] Takao Kotani and Mark van Schilfgaarde, Solid State Communications **121**, 7(2002)

[3] Mark van Schilfgaarde, Takao Kotani, and Sergey V. Faleev, in preparation.

Posters

Ab-Initio studies of structure and magnetic structure in YCo3H2.

X.Y. Cui^{1,2}, J. Liu^{1,3}, I. Morrison¹ and D.K. Ross¹

¹*Institute for Materials Research, University of Salford, Salford, M5 4WT, UK*

²*School of Physics, The University of Sydney, Sydney NSW 2006, Australia*

³*Department of Engineering Science, University of Oxford, Oxford, OX1 3PJ, UK*

Abstract

We present an ab-initio density functional study of magnetic phase transitions in the YCo3H2 system. The APW+lo method as employed in the WIEN2K code is used to predict the structure and electronic structure of this compound. Comparison is made with recent X-ray diffraction and magnetization studies. The calculations suggest that the YCo3H2 system is ferrimagnetic in character. Further, fixed spin moment calculations are used to predict and interpret magnetic phase transitions observed in externally applied magnetic fields.

A relativistic optimized potential method (OPM) for magnetic solids

H. Ebert and A. Perlov

Department Chemie/ Physikalische Chemie, University of Munich

Butenandtstr. 5-13, D-81377 Munchen, Germany

Abstract

The optimized potential method is based on a formulation of density functional theory that uses implicit functionals of the exchange-correlation energy with respect to the electronic density. For atoms there are already a number of corresponding investigations in the literature that turned out to be very fruitful. For solids on the other hand most work was done so far on insulators or semiconductors. For these systems the exchanged-only (exact exchange) version of the OPM led to band gaps in much better agreement with experiment than corresponding LDA-based calculations. For metallic ferromagnets, on the other hand, it was found by Kotani and Akai that an appropriate treatment of correlations have to be included. This way very satisfying results for magnetic properties could be obtained. In the case of the core hyperfine fields this seems to be caused primarily by the fact that the OPM leads to a self-interaction free exchange-correlation potential. Motivated by this success a relativistic formulation of the OPM has been developed that is formulated in terms of Green's function. The resulting current density OPM should in particular lead to better results for the spin-orbit induced orbital moments. Furthermore the developed formalism allows a very reliable implementation on the basis of the relativistic spin-polarized KKR-method. First results for the corresponding response functions and other related quantities will be presented.

Influence of correlation effects on the spin-resolved VB-XPS

J. Minar¹, H. Ebert¹, L. Chioncel², A. Lichtenstein², C. Nadai³, N. Brookes³

¹*Department Chemie/ Physikalische Chemie, University of Munich*

Butenandtstr. 5-13, D-81377 Munchen, Germany

jan.minar@cup.uni-muenchen.de,

²*University of Nijmegen, Toernooiveld 1, 6500 GL, Netherlands*

³*ESRF, Boite Postale 220, 38043 Grenoble Cedex, France*

Abstract

Calculations of valence band photoemission spectra on the basis of the local approximation to density functional theory have been very successfully in the past for many transition metal systems. However it is well known that correlation effects play for some materials a rather crucial role. Fortunately, it is often well justified to represent these by a local, i.e. site-diagonal self-energy. In this case correlation effects can straightforwardly be incorporated within calculations based on the one-step model of photoemission. This applies in particular for its spin-polarised relativistic version that allows to deal with the Fano-effect and magnetic dichroism.

Corresponding results will be presented for ferromagnetic Ni, for which the pure Fano-effect, that means the spin-polarisation of the photo-current due to spin-orbit coupling has been investigated. Results of investigations on the spontaneous spin-polarisation in the ground-state by means of spin-resolved VB-XPS will be presented for the half-metallic ferromagnet NiMnSb. Within these investigations correlation effects are accounted for via the electronic self-energy calculated by recently proposed LDA+DMFT scheme (DMFT: dynamical mean field theory). As it turn out, taking correlation effects into account the agreement between theory and experiment is significantly improved.

Electronic structure and bonding in (Mo,Cr)(Si,Al) alloys investigated by X-ray photoelectron spectroscopy and density-functional theory

D.P. Pankhurst¹, Z. Yuan², D. Nguyen-Manh^{1,3}, M.L. Abel², G. Shao², J.F. Watts², D.G. Pettifor¹ and P. Tsakiropolous².

¹ *Department of Materials, University of Oxford, Parks Road, Oxford, OX1 3PH, UK*

² *School of Engineering, University of Surrey, Guilford, Surrey GU2 7XH, UK*

³ *UKAEA Culham Division, Culham Science Centre, Abingdon OX14 3DB, UK*

We have investigated the electronic structure of (Mo,Cr)(Si,Al) alloys at a various range of composition using a combination of valence band X-ray photoelectron spectroscopy (VBXPS) and density-functional theory. We find good agreement between the experimental spectra and calculated total density of states. The observed differences between the VBXPS spectra for (Mo,Cr) Si₂, (Mo,Cr)₅Si₃ and (Mo,Cr)Si₃ are explained in term of the various hybridizations, involving Si p and Mo d states, induced by the structural topology. The changes observed in the VBXPS spectra for (Mo,Cr)(Si_(1-x)Al_x)₂ alloys with increasing Al concentration are explained by a series of structural transformations and a downward shift of the Fermi energy due to the reduced electron concentration. Using rigid band arguments we discuss how this leads to waekening of the covalent (Mo,Cr)-Si bonds and, hence, to increase ductility.

[1] D.A. Pankhurst, D. Nguyen-Manh and D.G. Pettifor, Phys. Rev. B., vol. 69, 075113 (2004)

[2] D.A. Pankhurst, Z. Yuan, D. Nguyen-Manh, Ml. Abel, G. Shao, J.F. Watts, D.G. Pettifor and P. Tsakiropolous, Phys. Rev. B., submitted (2004).

Ab initio modelling of high-k oxides.

Jacob Gavartin⁽¹⁾, Adam S. Foster⁽²⁾, Alexander Shluger⁽¹⁾

⁽¹⁾ *Department of Physics and Astronomy, University College London,
Gower Street WC1E 6BT, London, United Kingdom.*

⁽²⁾ *Laboratory of Physics, Helsinki University of Technology,
P.O. Box 1100, FIN-02015 HUT, Finland*

Abstract

The aggressive scaling of the Complementary Metal Oxide Semiconductor devices (CMOS) is an essential predicament for the success of the semiconductor industry. The key component of this miniaturization is an increase of a capacitance of the gate stack, which is achieved either through a reduction of the physical thickness of the gate dielectric (currently SiO₂) or/and through the use of new gate materials with higher dielectric constant. The latter approach has been a focus

of extensive academic and industrial research since the last publication of the *International Technology Roadmap for Semiconductors* around five years ago [1-3].

Remarkably, the problem of high-k dielectrics has opened both a large scope and a serious challenge for the electronic structure theory. On the one hand, density functional calculations stipulated significant progress in understanding complex interrelations between the stability of high-k films, their polymorphism and their dielectric response [3,4]. On the other hand, calculations exposed serious difficulties in predictive modelling of the interface growth, especially reliable values of the valence and conduction band offsets with silicon, properties of shallow intrinsic and extrinsic defects and the charge trapping associated with them.

We address some aspects of density functional modelling of defects in high-k oxides [4-7]. First, we discuss calculations of charged defects in bulk HfO₂ and at the Si/HfO₂ interfaces. Second, we overview the limitations of static calculations and consider how they can be instructively combined with the ab initio molecular dynamics to get new insight on the defect transformations in the material. Based on our extensive modelling, we discuss several pressing issues in high-k based CMOS technology:

1. Degradation of carrier mobility;
2. Fermi-level pinning and the threshold potential instability;
3. Role of controlled and uncontrolled impurities: water, hydrogen and various forms of nitrogen.

1. G. D. Wilk, and R. M. Wallace, and J. M. Anthony, *J. Appl. Phys.* **89**, 5243 (2001).
2. H. S. P. Wong, IBM. *J. Res. & Dev.*, **46**, 133 (2002).
3. M. Houssa (Editor), *High-k Gate Dielectrics*, IoP Publishing 2004.
4. A. L. Shluger, A. S. Foster, J. L. Gavartin, and P. V. Sushko, in *Nano and Giga Challenges in Microelectronics.*, eds. J. Greer, A. Korkin, and J. Labanowski, 151-222 (Elsevier, 2003).
5. A. S. Foster and F. Lopez Gejo and A. L. Shluger and R. M. Nieminen, *Phys. Rev. B*, **65**, 174117 (2002).
6. A. S. Foster and A. L. Shluger and R. M. Nieminen, *Phys. Rev. Lett.*, **89**, 225901 (2002).
7. J. L. Gavartin, A. S. Foster, G. I. Bersuker, A. L. Shluger, *J. Appl. Phys.* 2004 - submitted.

Dynamical properties of poly para-phenyl vinylene

G. Zheng, S. J. Clark, P. R. Tulip, S. Brand, and R. A. Abram
Department of Physics, University of Durham, DH1 3LE, United Kingdom

Abstract

It is well known that the vibrational spectroscopy of polyconjugated polymers is one of the tools for probing the structural and opto-electronic properties of materials in various phases both in pristine and doped states. The understanding of the relevant features requires a detailed knowledge of the phonon modes.

We present an ab initio investigation of the dynamical properties of the conjugated polymer poly para-phenylene vinylene (PPV) on both isolated chain and crystalline states within density functional perturbation theory (DFPT) framework implemented in the CASTEP plane wave code[1]. It is found that for an isolated PPV chain, most of the vibrational frequencies are previously assigned: the higher frequency phonon modes are due to the C-H stretch; the vinyl C-C stretch mode has a higher energy than the phenyl ring C-C stretch frequencies. Most of the out-of-plane modes are softer than those in-plane, since the former mainly involve bond-length and bond-angle alternation. However, there are several unidentified experimental modes, hence, calculations in the crystalline state allow us to assign the unidentified experimental modes. Also the results of the polarisability and permittivity of the material are reasonable when compared with typical axial values of conjugated polymers. Analyses of the first-order densities provide us insight into the behaviour of the electronic polarisabilities. Dynamical Born effective charges are more appropriate to be used for the study of the dynamics of the system. Notable differences are found in IR absorption spectra obtained for the PPV isolated and crystalline states which can be attributed to the differences to the crystalline packing effects which play a key role in the understanding of lattice dynamics of this system.

We acknowledge the funding by EPSCR grant no. GR/R56716/01.

1. M. D. Segall, Philip J. D. Lindan, M. J. Probert, C. J. Pickard, P. J. Hasnip, S. J. Clark, and M. C. Payne, *J. Phys.: Condens. Matter* 14, 2717 (2002).

Resolving Discrepancies Concerning the Enthalpy of Formation of Gallium Nitride

Michael C. Gibson, Stewart J. Clark, Stuart Brand, and Richard A. Abram
*Department of Physics, University of Durham,
DH13LE, United Kingdom*

Abstract

In recent years the study of gallium nitride and gallium nitride surfaces from first principles has become increasingly popular. The accurate study of surface reconstructions relies strongly on knowledge of the enthalpy of formation of the materials concerned as this determines the range of chemical potential for which the surface is stable. For example a phase diagram for hydrogen on GaN surfaces has recently been calculated [1] in which a value of 1.2 eV was taken as the enthalpy of formation of GaN. This was calculated from first principles using density functional theory (DFT) under the local density approximation and employing norm-conserving Kleinmann-Bylander pseudopotentials. This value has previously been considered reasonable as it is in agreement with commonly tabulated experimental values [2]. However more recent experimental results [3], as well as all-electron calculations [4], suggest the value may actually be in the region of 1.6 eV. In our own work presented here we employ projector reduced pseudopotentials [5] within in the CASTEP code [6] and arrive at a value in much better agreement with these results. We have employed these pseudopotentials to re-calculate the phase diagram for hydrogen on GaN surfaces, obtaining results very different to the previously published work [1].

References

- [1] C. G. Van de Walle and J. Neugebauer, *Phys. Rev. Lett.* **88**, 66103 (2002).
- [2] *CRC Handbook of Chemistry and Physics*, edited by David R. Lide (CRC Press, Boca Raton, Florida, 1992), 73rd ed., pp. 5-18.
- [3] M. R. Ranade, F. Tessier, A. Navrotsky, V. J. Leppert, S. H. Risbud, F. J. DiSalvo, and C. M. Balkas, *J. Phys. Chem. B* **104**, 4060 (2000).
- [4] M. Fuchs, J. L. F. Da Silva, C. Stampfl, J. Neugebauer, and M. Scheffler, *Phys. Rev. B* **65**, 245212 (2002).
- [5] Ming-Hsien Lee, *Advanced Pseudopotentials*, PhD Thesis (Cambridge).
- [6] CASTEP: M. D. Segall, P. J. Lindan, M. J. Probert, C. J. Pickard, P. J. Hasnip, S. J. Clark, and M. C. Payne, *J. Phys. : Cond. Matt.* **14**, 2717 (2002).

Generalised Kohn-Sham Theory

N. Carneiro

Abstract

Generalised Kohn-Sham (GKS) Theory [1,2] provides a variety of possible generalisations of the well-known Kohn-Sham implementation of density functional theory for the calculation of total energies of systems of interacting electrons such as molecules and solids. We present an assessment of various GKS schemes for a one-dimensional model semiconductor. Comparison is made with previous quantum Monte Carlo calculations [3] and with previous GW calculations within many-body perturbation theory for this system. Further GW calculations suggest a new GKS approach based on a GW formulation of the total energy. which will lead to a new version of density functional theory for efficient practical calculations which circumvents the problem of approximating the usual Kohn Sham exchange correlation energy functional.

- [1] A. Seidl, A. Görling, P. Vogl, J. A. Majewski and M. Levy; *Phys. Rev.* **B 53**, 3764 (1996)
- [2] P. Sánchez-Friera and R. W. Godby, *Phys. Rev. Lett.* **85**, 5611 (2000)
- [3] W. Knorr and R. W. Godby, *Phys. Rev. Lett.* **68**, 639 (1992)

Density functional theory study of fullerenes and endohedral fullerenes

S. M. Lee¹, D. Nguyen-Manh², G. A. D. Briggs¹, and D.G. Pettifor¹

¹*Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, UK*

²*UKAEA Culham Division, Culham Science Center, Oxfordshire OX14 3DB, UK*

Abstract

The atomic and electronic structures of fullerenes and endohedral fullerenes have been investigated using density functional theory. We have studied C₆₀, C₇₀ and C₈₂ fullerenes, and with an endohedral atom inside. A nitrogen atom in C₆₀ or C₇₀ was found to locate at the centre of the fullerene without chemical bonding to the fullerene, while a scandium atom in C₈₂ was stable at close to a hexagonal ring of fullerene cage. The relative stability of nine isomers of C₈₂ was changed dramatically by the encapsulation of a scandium atom. The applicability of these materials to quantum computing will be discussed.

Linear-scaling DFT calculations from empirical tight-binding to full plane-wave accuracy

T. Miyazaki¹, R. Choudhury², D. R. Bowler^{2,3} and M. J. Gillan²

¹*National Institute for Materials Science, 1-2-1 Sengen,*

Tsukuba, Ibaraki 305-0047, Japan

²*Dept of Physics and Astronomy, University College London,*

Gower Street, London WC1E 6BT, UK

³*ICYS, National Institute for Materials Science, 1-1 Namiki,*

Tsukuba, Ibaraki 305-0044, Japan

Abstract

Density functional theory (DFT) has become a standard tool for modelling materials. But conventional methods are very inefficient for large complex systems, because the memory requirements scale as the square of the number of atoms, and the cpu requirement as the cube of the number of atoms. We report recent progress in the development of the CONQUEST code [1-6], which performs O(N) (linear scaling) DFT calculations on parallel computers, and has a demonstrated ability to handle systems of many thousands of atoms [3- 5]. The code is based on the strategy of minimizing the total energy with respect to the Kohn-Sham density matrix, and the practical techniques for implementing this strategy will be briefly summarized. The code can be run at different levels of precision, ranging from empirical

tight-binding, through ab initio tight-binding, to full plane-wave precision, and the way in which this is achieved will be outlined. Illustrations will be given of very recent practical CONQUEST calculations on semiconductor surfaces. The ability to run at all levels of precision up to plane-wave precision will be illustrated by results on the reconstructed Si (001) surface, and the ability handle large complex systems will be illustrated by preliminary results on three-dimensional reconstructions of Ge overlayers on Si (001) requiring DFT calculations on systems of over 3000 atoms.

- [1] E. Hernandez, M. J. Gillan, Phys. Rev. B, 51, 10157 (1995)
- [2] E. Hernandez, M. J. Gillan, C. M. Goringe, Phys. Rev. B, 53, 7147 (1996)
- [3] C. M. Goringe, E. Hernandez, M. J. Gillan, I. J. Bush, Comput. Phys. Commun., 102, 1 (1997)
- [4] D. R. Bowler, T. Miyazaki, M. J. Gillan, Comput. Phys. Commun., 137, 255 (2001)
- [5] D. R. Bowler, T. Miyazaki, M. J. Gillan, J. Phys. Condens. Matter, 14, 2781 (2002)
- [6] T. Miyazaki, D. R. Bowler, R. Choudhury, M. J. Gillan, J. Chem. Phys., 121, 6186 (2004)

Molecular dynamic calculations of GaN thermodynamic properties

V. TIMON, S. BRAND, S. J. CLARK AND R. A. ABRAM

Department of Physics, University of Durham, South Road, Durham DH1 3LE, UK

Abstract

The thermal expansion coefficients of (wurtzite) GaN at low and intermediate temperature are known and experimental data are available [1, 2] but at high temperatures it doesn't happened. Also due to experimental difficulties the GaN melting point is not reliably known and only a few theoretical studies have been made [3]. Based on the NPT (isothermal- isobaric) ensemble, we have carried out a molecular dynamic simulation in the DFT framework to obtain the coefficients of thermal expansion for a single solid phase of GaN from 0 K to high temperatures. An estimation of the melting point temperature T_m at normal pressure (1 Atm) has been made.

- [1] R. R. Reeber, K. Wang , Journal of Materials Research 15, 40-44 (2000).
- [2] M. Leszczynski, T. Suski, H. Teisseyre, P. Perlin, I. Grzegory, J. Jun, S. Porowski, T. D. Moustakas, Journal of Applied Physics 76(8), 4909 (1994).
- [3] K. Harafuji, T. Tsuchiya, and K. Kawamura, Phys. Stat. Sol. (c), 176, No. 7, 2420-2423 (2003).

(E-mail: v.t.salinero@durham.ac.uk)

Self-interaction correction (SIC) in multiple-scattering theory

Martin Lüders¹, Arthur Ernst², Markus Däne³, Džidka Szotek¹, Axel Svane⁴,
Diemo Ködderitzsch^{3,1}, Wolfram Hergert³, Balazs L. Györfy⁵, Walter Temmerman¹

¹ *Daresbury Laboratory, Daresbury, Warrington, UK*

² *Max Planck Institut für Mikrostrukturphysik, Halle, Germany*

³ *Fachbereich Physik, Martin-Luther-Universität Halle-Wittenberg, Germany 06099*

⁴ *Institute of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus, Denmark*

⁵ *H.H. Wills Physics Laboratory, University of Bristol, UK*

Abstract

We propose a simplified version of self-interaction corrected local spin-density (SIC-LSD) approximation, based on multiple scattering theory, which implements self-interaction correction locally, within the KKR method. The multiple scattering aspect of this new SIC-LSD method allows for the description of crystal potentials which vary from site to site in a random fashion and the calculation of physical quantities averaged over ensembles of such potentials using the coherent potential approximation (CPA). This facilitates applications of the SIC to alloys and pseudoalloys which could describe disordered local moment systems, as well as intermediate valences. As a demonstration of the method, we study the well-known α - γ phase transition in Ce, where we also explain how SIC operates in terms of multiple scattering theory.

Ab Initio Calculation of Structure and Phonon Vibration in Ices

Tsogbadrakh Namsrai and Ian Morrison

Joule Physics Laboratory, Institute for Materials Research, The University of Salford, Salford, M5 4WT, UK.

Abstract

First principles calculations of ice, and orientational defects in ice have been performed within a framework of DFT [1, 2] using both the plane wave pseudopotential method (CASTEP) [3] and the flexible LCAO method (SIESTA)[4].

The structure, electronic structure and phonon frequencies of ice Ih are calculated using both methods are compared with each other and with experiment. Calculations using the less computationally demanding SIESTA and the DZP basis set are shown to be quantitative. Within this computational framework the sensitivity to calculated structural properties including zero point vibrations is estimated and shown to be small even though changes in zero point energies account for a significant fraction of the energy of formation [5].

Further, we have performed calculations of L and D orientational defects in ice within a supercell approach. In an orientational defect the ice rules [6] are broken such that in a D - defect two hydrogens lie between nearest neighbour oxygens, and in a L - defect zero hydrogens lie between nearest neighbour hydrogens [7]. The dynamics of these defects forms the basis of transport properties in ice. The structure and electronic structure of these

defects in a supercell containing both defect types is calculated and compared to the idealised geometry.

Acknowledgement

We thank the ORS Awards Scheme of United Kingdom Scholarships for International Research Students and the Salford's University for the financial support during this work.

References

- [1] P. Hohenberg and W. Kohn, *Phys. Rev.*, **1964**, (136), p. B864-B871
- [2] W. Kohn and L.J. Sham, *Phys. Rev.*, **1965**, (140), p. A1133 - A1138
- [3] M. D. Segall and et al., *J. Phys: Condens. Matter*, **2002**, (14), p. 2717 - 2744
- [4] Jose M. Soler and et al., *J. Phys: Condens. Matter*, **2002**, (14), p. 2745 - 2779
- [5] Tsogbadrakh N. and et al., *J. Chem. Phys.*, (to be published)
- [6] Bernal J. D. and Fowler R. H., *J. Chem. Phys.*, **1933**, (1), p. 518 - 548
- [7] Niels Bjerrum, *Science*, **1952**, (115), p. 385 - 390

Electronic and magnetic properties of the surfaces of hole-doped manganites

H. Zenia¹, G. A. Gehring¹, W. M. Temmerman²

¹*Department of Physics and Astronomy, University of Sheffield, UK*

²*Daresbury Laboratory, Daresbury, UK*

Abstract

The electronic structure and magnetic phases of hole-doped manganite surfaces are investigated in a model calculation. The model incorporates the kinetic energy of the e_g electrons, their coupling through Hund's rule to the core-like $S=3/2$ t_{2g} spin and the superexchange interaction between the latter [1]. The anisotropy of the transfer integrals leads to important physics at the surface/interface. There is only one finite transfer integral between planes connecting the $3z^2 - r^2$ orbitals. The latter is then much more sensitive to the presence of the surface, which is perpendicular to the z -axis, than the $x^2 - y^2$ orbital. As a result, at the surface, the projected DOS for the $x^2 - y^2$ orbital is broader than that of $3z^2 - r^2$ orbital. The effect of the broken cubic symmetry at the surface is to favour the occupation of the $x^2 - y^2$ orbital while in the bulk both orbitals are equally occupied. We added a shift Δ [2] to the onsite energy at the surface in order to mimic the effects of the broken symmetry and of the changes in the type, number and distance to neighbouring atoms at surfaces or

interfaces with other materials. We looked at negative values of Δ added to one orbital or both while the bulk levels are taken as the reference for the energies. We found that the most favourable scenario is to add a small shift Δ to the $3z^2 - r^2$ to enhance the stability of the ferromagnetic (FM) coupling between surface and bulk. A larger Δ would deplete the other orbital and thus would disfavour in-plane FM coupling at the surface which would be detrimental to the tunnelling magnetoresistance (TMR) of manganite-based junctions. This could be achieved by growing the manganite film on an insulating oxide with a lattice parameter such that the distance between the interface Mn ion and the oxygen ion in the next layer is slightly larger than the Mn-O bond in the bulk of the manganite.

References

- [1] Jeroen van den Brink and Daniel Khomskii, Phys. Rev. Lett. **82**, 1016 (1999).
- [2] M. J. Calderón, L. Brey, and F. Guinea, Phys. Rev. B **60**, 6698(1999).

Electronic structure of UO_2 and its point defects: an LSDA+U approach

Younsuk Yun¹, Hanchul Kim², Kwangheon Park¹

¹ *Kyung Hee University, Department of Nuclear Engineering, Suwon, 449-701, Korea*

² *Korea Research Institute of Standard and Science, P.O.Box 102, Yuseong, Daejeon, 305-600, Korea*

Abstract

We have investigated the electronic structure of UO_2 using the density functional theory (DFT) calculations within the local spin-density approximation (LSDA)+U approach as implemented in the Vienna Ab initio Simulation Package (VASP). For the bulk UO_2 , the electronic structures from both the LDA and the LSDA+U approaches are compared in order to elucidate the effect of correlation. The calculated cohesive properties of UO_2 are compared with available experimented data. For different point defects in UO_2 , the formation energy and the defect-induced changes in the electronic structure were investigated.

Ab Initio Modelling of Complex Hydrogen-bonded Structures using Density Functional Theory

Duncan Riley, Ian Morrison

Institute of Materials Research, University of Salford, England.

Abstract

This project is looking at the feasibility and accuracy of modelling complex hydrogen bonded systems to obtain information on structure and vibrational spectra. The first system looked at was N-Hydroxyphthalimide (NHP) crystallized in its yellow form. This was modelled using the CASTEP [1] code, a Density Functional Theory (DFT) program using plane wave basis sets and pseudopotentials. The first phonon spectra were calculated in the harmonic approximation using both the finite difference approach and linear response theory. The Structure of yellow NHP, calculated using geometry optimisation, will be shown as well as comparisons of the density of phonon states in the crystalline solid obtained using finite difference and linear response methods. Result from linear response analysed by the a-Climax program [2] will be displayed and compared to experimental neutron scattering data.

[1] M D Segall, Philip J D Lindan, M J Probert, C J Pickard, P J Hasnip, S J Clark and M C Payne, *J. Phys: Condensed Matter* 14, 2002

[2] A. J. Ramirez-Cuesta, *Computer Physics Communications*, Vol. 157/3 Pp 226-238, 2004
d.j.riley@pgr.salford.ac.uk

GW and SIC implementation in the KKR method

A. Ernst, V. Dugaev, P. Bruno

Max-Planck-Institut für Mikrostrukturphysik Halle(Saale), 06120, Germany

M. Lüders, W.M. Temmerman, Z. Szotek

Daresbury Laboratory, Warrington WA4 4AD, United Kingdom

B. Györfy

HH Wills Physics Laboratory, University of Bristol, UK

Abstract

The *ab-initio* study of semiconductors and insulators as well as systems with strong localized electrons entails great difficulties involved by the treatment of excitation energies and many-body effects. The most successful first principles method, the density functional theory (DFT) within the local spin density approximation (LSDA), is designed for ground state properties and can not provide proper description of band structure of semiconductors and insulators. If some localized electrons are present in the system, like *3d*-electrons in the transition metal oxides, the local density approximation can be essentially improved by so-called self-interaction correction (SIC). In this approximation the self-iterations of single particle charges, which are present in the LSDA, can be canceled out for the localized electrons. However, the self-interaction correction within the LSDA is still not sufficient for proper description of the excitation energies and band-gaps. It is possible to do accurately from first principles by solving the Hedin's set of equations for the full Green's function. The implementation of this formalism is very difficult, one neglects commonly the vertex correction (the random-phase approximation), and the self-energy is calculated in this case within so-called GW approximation. The non-self-consistent GW approximation was successfully

implemented within several first-principle methods, but most of existing implementations are generally designed for systems with delocalized (fast) electrons.

Here we present a general *ab-initio* approach designed for the study of electronic properties of solids, in which on base of the Korringa-Kohn-Rostoker (KKR) method we implemented the self-interaction correction for strong-localized electrons and the non-self-consistent GW approximation for the inclusion of many-body effects. We illustrate our approach on the electronic structure study of some semiconductors and transitions metal.

Self-interaction correction (SIC) in multiple-scattering theory

Martin Lüders¹, Arthur Ernst², Markus Däne³, Dzikka Szotek¹, Axel Svane⁴,
Diemo Ködderitzsch^{3,1}, Wolfram Hergert³, Balazs L. Györfy⁵, Walter Temmerman¹

¹ *Daresbury Laboratory, Daresbury, Warrington, UK*

² *Max Planck Institut für Mikrostrukturphysik, Halle, Germany*

³ *Fachbereich Physik, Martin-Luther-Universität Halle-Wittenberg, Germany 06099*

⁴ *Institute of Physics and Astronomy, University of Aarhus, DK-8000 Aarhus, Denmark*

⁵ *H.H. Wills Physics Laboratory, University of Bristol, UK*

Abstract

We propose a simplified version of self-interaction corrected local spin-density (SIC-LSD) approximation, based on multiple scattering theory, which implements self-interaction correction locally, within the KKR method. The multiple scattering aspect of this new SIC-LSD method allows for the description of crystal potentials which vary from site to site in a random fashion and the calculation of physical quantities averaged over ensembles of such potentials using the coherent potential approximation (CPA). This facilitates applications of the SIC to alloys and pseudoalloys which could describe disordered local moment systems, as well as intermediate valences. As a demonstration of the method, we study the well-known α - γ phase transition in Ce, where we also explain how SIC operates in terms of multiple scattering theory.

Ab-Initio Theory of Resonant X-ray Scattering in Rare Earth Materials

P. Strange¹, M. Horne¹, E. Arola^{1,2}, S.J. Stockton¹,
B. Zarychta¹, H. Winter³, Z. Szotek⁴ and W.M. Temmerman⁴

¹ *School of Chemistry and Physics, Keele University, Staffordshire ST5 5BG, UK*

² *Optoelectronics Research Centre, Tampere University of Technology, P.O. Box 692,
FIN-33101, Tampere, Finland.*

³ *INFP, Forschungszentrum Karlsruhe GmbH, Postfach 3640, D-76021 Karlsruhe, Germany*

⁴ *Daresbury Laboratory, Daresbury, Warrington, WA4 4AD, Cheshire, UK*

Abstract

A first-principles theory of resonant magnetic scattering of x rays is presented. The scattering amplitudes are calculated using a standard time-dependent perturbation theory to second order in the electron-photon interaction vertex. In order to calculate the cross section reliably an accurate description of the electronic states in the material under investigation is required and this is provided by the density functional theory (DFT) employing the Local Spin Density Approximation combined with the self-interaction corrections (SIC-LSD). The magnetic x-ray resonant scattering (MXRS) theory has been implemented in the framework of the relativistic spin-polarized LMTO-ASA band structure calculation method. The theory is illustrated with two applications. Firstly we will show a series of calculations for praseodymium that display the sensitivity of MXRS to the symmetry of the occupied f -states. Secondly we will make direct contact with experiment with a calculation for holmium that can be compared with recent asymmetry ratio measurements from the XMAS beamline at ESRF.

First-principles calculation of the effects of structural relaxation on the optical absorption spectra of ruby and alexandrite

Kazuyoshi Ogasawara, Tomomi Sasaki, Rie Taniguchi, Shinta Watanabe, Takugo Ishii

Department of Chemistry, Kwansai Gakuin University,

2-1 Gakuen, Sanda, Hyogo 669-1337, JAPAN

E-mail: ogasawara@ksc.kwansei.ac.jp

Abstract

The d-d transitions of transition-metal (TM) ions in ionic crystals have been utilized as solid state lasers such as ruby (chromium doped alumina) and alexandrite (chromium doped chrysoberyl). The optical spectra of these materials have been generally analyzed based on the semiempirical ligand field theory. In this approach the parameters are determined by fitting the theoretical multiplet energies to the experimental ones under appropriate assignment of the observed peaks. However, additional parameters are required for consideration of covalency and the number of parameters increases for lower-symmetry systems. Therefore, establishment of non-empirical computational approach is quite important for unambiguous analysis of materials with complicated local structure. In order to establish a nonempirical computational approach for the optical spectra for impurity transition-metal ions or also rare-earth ions, we have recently developed a first-principles relativistic configuration-interaction (CI) calculation program, in which many-electron wave functions are obtained as linear combination of relativistic Slater determinants by direct diagonalization of the many-electron Dirac Hamiltonian. The transition probabilities between the eigenstates are calculated directly using the explicit many-electron wave functions. The TM L_{2,3}-edge X-ray absorption near-edge structures (XANES) for some TM compounds have been calculated by this approach and excellent agreement with the experimental spectra was obtained [1]. For the analysis of optical spectra of impurity ions, it is also important to consider the effects of structural relaxation around them. Therefore, in this work, we have investigated the effects of structural relaxation around the chromium ions on the absorption spectra of ruby and alexandrite by combining the structural optimization using CASTEP code and first-

principles CI calculations using model clusters composed of 63 atoms (ruby) or 60 atoms (alexandrite). In the case of ruby, the reproducibility of the characteristic anisotropy of the absorption spectra was improved both qualitatively and quantitatively by consideration of the structural relaxation. However, in the case of alexandrite, the reproducibility became worse even qualitatively by consideration of the structural relaxation, indicating the difficulty in predicting structural relaxation for lower- symmetry systems.

[1] K. Ogasawara et al., Phys. Rev B 64, 115413 (2001).

First Principles Study of the Kondo Effect

Andrew. I. Duff, J. F. Annett

H. H. Wills Physics Laboratory, University of Bristol, Tyndall Ave, BS8 1TL.

Abstract

The Kondo effect is a non-perturbative many-body phenomenon arising for a single magnetic impurity atom in a metal. First principles theory based on DFT LDA does not include the quantum dynamics of the impurity spin, and hence the Kondo effect.

Here the Kondo effect is to be studied from 1st principles. The starting point is DFT calculations of an atom embedded in jellium. The self consistent approach used by Manninen et al [1] is used to look for a magnetic solutions of the system in terms of the two model parameters: the charge of the ion and the charge density of the jellium. In particular, the system of an Iron atom in jellium is studied. Magnetic and non-magnetic solutions are found, and the difference in energy between the solutions is calculated as a function of jellium density. The magnetic moment is also calculated for the range of jellium densities at which it occurs.

The Local Density Approximation (LDA) neglects the quantum correlations which lead to the Kondo effect. To go beyond LDA we are using a Quantum Monte Carlo approach

[1] M. Manninen, R. Nieminen, P. Hautojarvi, and J. Arponen, Phys. Rev. B 12, 4012 (1975)